

Accessing Higher Vibrational States of He_2^+ through Multi-Step ExcitationM. Holdener¹, F. Merkt^{1*}¹Departement of Chemistry and Applied Biosciences, ETH Zurich

The precise measurement of highly excited vibrational states of small molecular systems such as H_2^+ , H_2 and He_2^+ is of significant interest as benchmark to *ab-initio* quantum-chemical calculations [1]. In the case of He_2^+ , only the first few vibrational levels of the electronic ground state $X^+ \ ^2\Sigma_u^+$ were precisely measured [2-3]. In this poster presentation, we propose a method to access the higher vibrational states of He_2^+ $X^+ \ ^2\Sigma_u^+$ using a multi-step excitation scheme.

Our approach involves the production of a molecular beam of He_2 in the long-lived metastable $a \ ^3\Sigma_u^+$ state [4] through an electric discharge. We then utilize a high-intensity laser to promote the system to the electronic state $c \ ^3\Sigma_g^+$ in an excited vibrational level with v in the range 3-5. This state predominantly decays radiatively to the $a \ ^3\Sigma_u^+$ state with $v' = v$ because of favorable Frank Condon factors.

A second laser is then employed to induce a transition from the vibrationally excited metastable state $a \ ^3\Sigma_u^+(v')$ to the ion state $X^+ \ ^2\Sigma_u^+(v^+ = v')$. This method offers the prospect of studying the structure and dynamics of highly excited vibrational levels of He_2^+ that are typically challenging to access.

[1] Dávid Ferenc, Vladimir I. Korobov, and Edit Mátyus, *Phys. Rev. Lett.*, **2020**, 125, 213001.

[2] Luca Semeria, Paul Jansen, Gian-Marco Camenisch, Federico Mellini, Hansjürg Schmutz, and Frédéric Merkt, *Phys. Rev. Lett.*, **2020**, 124, 213001.

[3] Paul Jansen, Luca Semeria and Frédéric Merkt, *J. Chem. Phys.*, **2018**, 149, 154302.

[4] Cary F. Chabalowski, James O. Jensen, David R. Yarkony, Byron H. Lengsfeld, *J. Chem. Phys.*, **1989**, 90, 2504-2512.